USGS

WATER RESOURCES RESEARCH GRANT PROPOSAL

Title: Quantifying Urban Non-point Sources of Lead for use in TMDL Computations

Focus Categories: NPP, WQL, M&P

Keywords: lead, roof runoff, urban runoff, non-point source pollution, total maximum daily load

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Non-Federal Funds Pledged: $44,165

Principal Investigator: Dr. Laura J. Steinberg, Tulane University, Louisiana Water Resources Research Institute

Congressional District: 2nd District of Louisiana

Statement of Critical Regional Water Problems

This project addresses the critical water problem stemming from lead contamination of Louisiana's rivers, streams, and lakes. The 1998 State of Louisiana Water Quality Management Plan, Water Quality Inventory Section 305(b) reports that lead is a significant source of water quality degradation in the state: lead accounted for impairment of 104, 248 acres of Louisiana lakes and 2,475 miles of Louisiana rivers and streams. Identifying and quantifying non-point sources of this contamination is imperative if the degradation is to be controlled and water quality improved to meet acceptable water-use levels.

Statement of the Results, Benefits, or Information to be Gained:

In developing total maximum daily loads (TMDLs) for these water bodies, it is imperative that non-point sources of lead be identified and quantified so that:

1. realistic allocations of lead load can be made, and

2. appropriate non-point source mitigation policy can be formulated.

Recent evidence indicates that runoff from leaded-paint exterior walls and rooftops constitute a large part of the lead presently described as "other urban runoff" in the Lousisiana Dept. of Environmental Quality's tabulations of "suspected sources of non-support of designated uses'. This project will both *qualitatively* assess the contribution of rooftops and leaded-paint walls to urban non-point sources of lead, and *quantatively* model lead concentrations in this runoff as a regression function of rainfall and building characteristics. The result will be a modeling tool that decision-makers can use to estimate the amount of lead released from rooftop and exterior wall runoff during rainfall events. This tool will aid the decision-makers in making load allocations and prioritizing non-point source mitigation efforts.

The model will be developed for New Orleans, however, validation tests can be undertaken in a post-project phase to determine its applicability to other regions of the state and nation.

Quantifying Urban Non-point Sources of Lead for use in TMDL Computations

Nature, Scope and Objectives of the Research

Louisiana has designated 104, 248 acres of lakes and 2,475 miles of rivers and streams as being environmentally degraded due to lead contamination (Louisiana Dept. of Environmental Quality, 1998). In the development of total maximum daily loads (TMDLs) , as mandated by the Clean Water Act, the state must set maximum limits for the discharge of pollutants, accounting for the non-point sources of the contaminants. Recent evidence has indicated that two sources may be contributing to non-point source loadings of lead, yet the effect of these sources on water quality has not been adequately investigated. These sources are lead from roof runoff and lead from rain running off exterior walls with lead paint facades. These sources will be investigated and modeled in this project. More specifically, the objectives of this work are:

1. To *qualitatively* assess the potential for residential home rooftops and exterior walls to serve as non-point sources of lead in urban runoff.

2. To formulate and parameterize *quantitative* regression models which predict the concentration of lead in runoff from residential home rooftops and exterior walls as a function of rainfall and building characteristics.

3. To measure the size distribution of the lead particulates in the runoff as input toa fate and transport model for lead emanating from these two sources, to be developed in a future project.1

As states, including Louisaina, develop total maximum daily loads (TMDL's), and then seek to allocate these loads to point and non-point sources, it is imperative that all sources of pollutants be identified, and that predictions be made of stormwater runoff concentrations of these pollutants. For point sources which regulate their own discharges, such as industrial facilities, such predictions may be easy to make. Non-point source loadings, however, are far more difficult to estimate, since they are erratic and are controlled by rainfall and other environmental conditions which may not be measured. Completion of this study will aid TMDL development in the following ways:

1. provide *quantitative evidence* that rooftops and homes are an important subset of urban runoff now classified as “other” in state TMDL development documents.

2. provide a *method* for estimating the quantity of lead running off from rooftops and homes, both in the dissolved form and the particulate form, and under various rainfall scenarios.

3. provide a size distribution of lead particles emanating from these sources for later use in urban runoff models.

All data will be collected in the City of New Orleans, where very high lead concentrations in the soil have been found (Mielke et al.1999). The roof runoff will be collected from the type of residential roof most commonly found in New Orleans – an asphalt-shingle roof, and the resulting roof runoff model will be applicable to this type of roof only (not to metal roofs, or other roof materials). These roofs are not expected to generate lead themselves, but to serve as storage reservoirs for lead particles which are deposited as wet or dry deposition. The source of these lead particles could be industrial air emissions, vehicle emissions, soil lead particles resuspended by strong winds or human activities, or leaded paint particles released into the air.

Wall runoff will be collected from private homes with leaded paint facades. It is expected that the quantity of wall runoff will be small for rain events with low intensities, as the roof will collect most of the water. However, with high wind speeds or driving rains, it is expected that rain will wash down the sides of the buildings, generating lead loadings into the water. We expect that the greater the intensity of the rainfall, the more force will be exerted on the lead paint, and the more lead particles may be dislodged from the paint and enter the wash water. We also expect that pH may play a role in determining lead concentrations in the wall runoff because strong acids are capable of dissolving lead.

Much of the lead will be in particulate form – we are interested in determining the size and density of these lead particles as input to a lead fate and transport model, to be developed in a later project. Smaller particles of lead are likely to be washed along into the drainage system of the area, and, in New Orleans, end up as sediment in the bottom of the drainage canals. These will serve as a constant, low level source of lead to the canal waters, as well as being a constant threat to the water quality of Lake Ponchartrain when hurricanes or very high flows flush out the canals. Larger particles are likely to settle out into the soil before reaching the drainage system. These larger particles may become incorporated in the soil matrix, creating hazardous conditions for passerby’s (especially children playing) as well contributing to dissolved lead concentrations in runoff water over the soil.

Methods, Procedures, and Facilities

<u>Selection of homes for the study:</u>

Paint chips will be collected from homes in New Orleans located in neighborhoods of New Orleans which have previously been identified as having high lead levels in soils (Mielke et al, 1999). Homeowners will be asked for permission to take sample paint chips from their homes. In anticipation of possible resistance from homeowners to participate in this study, we will work through neighborhood associations for introduction to individuals who are likely to be receptive to working with us. Paint chips will be analyzed according to the procedure in Section 12.4 below; a random sample of 10 homes with high lead content (greater than 1000 mg/kg) will be retained for the study. Six of these homes will be used for roof runoff collection and ten for wall runoff collection. In addition, two homes with low lead content (less than 100 mg/kg) in the same neighborhoods will be used as controls for the wall runoff sampling.

Roof runoff collection procedure:

Three homes in the study group will be outfitted with PVC rooftop rain gutters which will serve as the rooftop runoff collection system. The selection of PVC material ensures that all lead in the rooftop runoff comes from rooftop drainage and not from rain gutters. Following Yaziz (1989), these gutters will drain into five sample bottles. Each sample bottle contains a ping-pong ball which will block the opening of the bottle as it fills up. In this way, rooftop runoff will be collected for the “first flush” and then for four distinct periods afterwards. The length of these periods will depend on the rainfall intensity and the amount of roof square-footage which drains into the rain gutter.

Sampling will proceed during 10 rainfall events throughout the year. Just prior to the rain event, we will connect four pre-conditioned 200 ml polyethylene bottles to the rooftop runoff collection system. The pre-conditioning will consist of cleaning the bottles with hydrochloric acid and deionized water in a 1:6 ratio and soaking overnight. One pre-conditioned 200mL polyethylene bottle with funnel for rainfall collection will be placed in a site open to direct rainfall as a control. The rainfall intensity at each home will be measured with an in-place rain gauge. Intensity will be measured in 15 minute intervals by recording the amount of rainfall collected in that time period. The rain gauge provides precision on the order of +/- 1 millimeter.

In addition to the three homes outfitted with PVC rain gutters, we will also sample roof runoff from three other homes in the study group which have in-place metal rain gutters. These gutters also will be connected to the five-bottle sampling system described above. In this way, we will be able to evaluate the individual contribution of lead from metal gutters to lead concentrations in urban runoff.

Exterior Wall Collection Procedure:

Sampling will proceed during 20 rainfall events throughout the year. A random sample of five homes from the ten homes identified earlier with high lead content and one of the low-lead content homes will be sampled during each rain event. People-power on the project is not sufficient to sample more than six homes during any single rain event. Aluminum foil fashioned into a trough will be placed along the edge of the building during the rainstorm and held there until 50 ml. of rainwater have been collected. The rainwater will then immediately be transferred to the pre-conditioned polyethylene bottles.

Pretreatment of Runoff Samples

Immediately after collection, the samples will be measured to determine pH and temperature. They will then be divided in half and one sub-sample wll be filtered through a 0.2 micron filter. Both samples will then be acidified with sufficient HCl to bring the pH down to 2. It is assumed that this acidification will be sufficient to dissolve all lead particles. Particulate lead concentration will be quantified by subtracting the total lead value of the unfiltered portion from the lead concentration in the filtered portion. After acidification, samples will occasionally be agitated while stored at 4+/- 2 degrees C until the lead analysis is performed.

Paint chips

Paint chips from selected buildings will be collected for total lead analysis by removing loose, pealing pain with a glass rod. They will be transported to the laboratory in wide-mouthed 200 ml polyethylene bottles preconditioned with HCl and deionized water. Acid digestion of the chips will be performed in a CEM microwave. Prior to placement in the microwave vessel, a 0.5 g sample will be cut from the chip and placed in 10 ml of HNO_3. Micro-waving will proceed in 4 stages, with pressure increasing from 20 psi to 80 psi. Following complete digestion, the sample will be transferred to the ICP-MS for lead analysis.

Lead Analysis

A Finnegan ICP-MS maintained with magnetic sector instrument will be used to perform all lead analyses. The precision of this ICP-MS is at the level of parts per trillion. The analysis will be performed under contract by Tulane's Coordinated Instrumentation Facility (CIF) by professional chemists experienced and knowledgable about the use of ICP-MS equipment. All necessary precautions will be taken by the CIF to avoid inadvertant contamination of the samples.

Tulane Coordinated Instrumentation Facility

The Coordinated Instrumentation Facility is a Tulane-wide department operating under the Tulane Office of Research for the management of shared research equipment. CIF professional laboratory personnel maintain and operate the analytical equipment and assist researchers with method development and data collection. The CIF operates complete microscopy, inorganic, and organic laboratories. In addition to the ICP-MS to be used in this project, the CIF's inorganic analysis laboratory also includes an x-ray fluorescence spectrometer, scanning electron microscope, and graphite furnace atomic absorption spectrometer for inorganic analyses.

Particle Size Analysis

The filtered particles collected during the sample pretreatment (see Section 12.4) will be filtered through progressively smaller pores, beginning with geotechnical sieves, and proceeding through a set of cloth or paper filters. At each filtration step, the rejected mass will be weighed to yield the mass of particles in each size category.

Regression Modeling

Quantitative modeling of the lead data will be accomplished using regression models. It is anticipated that two sets of models will be generated: one for predicting rooftop runoff concentration, *LR*, and one for predicting wall runoff concentrations, *LW*. It is anticipated that the model forms will be linear, although non-linear regression can be employed if necessary. For the rooftop runoff model, the initial formulation of the model will be:

$$LR_T = B_0 + B_1 LS + B_2 INT + B_3 pH + e \qquad \text{Eq. 1}$$

where: LR_T is the total lead concentration in the runoff , *LS* is the number of days since the last rainstorm, *INT* is the intensity of the rainstorm, *pH* is the pH of the rainwater, the ***B****'s* are the regression parameters, and *e* is the model error. The regression procedure is used to determine the best value of the ***B***'s, using the criterion that the squared errors of the predicted value (LR_T) are minimized (the least squares regression). A second, similar equation for dissolved lead, LR_D, will also be formulated and solved using the same regression procedure:

$$LR_D = B_0 + B_1 LS + B_2 INT + B_3 pH + e \qquad \text{Eq. 2}$$

Linear regression models of a similar form will be developed for the wall runoff data. The independent variable, *LW*, will be modeled as:

$$LW_T = B_0 + B_1 LS + B_2 INT + B_3 pH + B_4 PC + e \qquad \text{Eq. 3}$$

where**:** LW_T is the total lead concentration in the wall runoff and *PC* is the concentration of the lead in the paint chip. An exactly analgous equation will be created for the dissolved lead in the wall runoff, LW_D:

$$LW_D = B_0 + B_1 LS + B_2 INT + B_3 pH + B_4 PC + e \qquad \text{Eq. 4}$$

These initial formulations will be tested for the standard violations of the assumptions of the linear regression model. Specifically, the models will be tested for violations of non-normality, non-constant variance, and serial correlation. Transformations to the data to correct these problems, should they arise, will be implemented. These might include log transformations, power transformations, or differencing of the data. If required to improve model fit, non-linear regression will be employed. All modeling will be performed with the statistical software “SPLUS 2000” which the PI is quite familiar with.

Related Research

Numerous publications have documented the presence of lead in urban runoff (Ellis 1977; Whipple and Humter, 1981; USEPA, 1983; Cole et al.1984; Flores-Rodriguez et al. 1993, Martin 1995, Tsihrintzis and Hamid 1998). Much of this lead has been attributed to road and highway runoff (Kerri 1985, Watershed Protection Techniques, 1994). While this is certainly an important source (although far less so than in 1985, when leaded gasoline was still widely available), recent studies have indicated that residential homes may also be important sources of urban lead pollution.

Yaziz et al. (1989) studied lead concentrations emanating from tile and galvanized iron roofs in Malaysia. He found average levels of “first flush” lead in the roof runoff of 235 micrograms/l for the galvanized iron roof and 102 micrograms/l for the tile roof. Over the course of the rain events, the galvanized roof and tile roof concentrations as high as 254 μg/l and 271 μg/l, respectively, were recorded. The average rainwater concentration over several storm events was 200 μg/l, but the concentration range reported is so large (40 μg/l to 520 μg/l) that a comparison to the average values given for the runoff concentrations is meaningless.

Good (1993) studied roof runoff lead concentrations at a Washington state sawmill whose runoff flows directly into the sea. He found that heavy metal concentrations of copper, lead, and zinc all exceeded the EPA standards for disharge into marine waters. Sampling was done for several different roof types (galvanized metal, roofing paper and tar, and anodized aluminum). Even the tar roof, which is not expected to contain any lead in its constituent material, produced unacceptably high concentrations of lead. Good collected samples at the beginning of the rain event, and then again three hours later. He found that the metal concentrations were generally lower after the “first flush” and theorized that this resulted from the early removal of easily-dislodged particles containing the metals.

Davis and Burns (1999) measured concentrations of lead in roof runoff in Prince George County, Maryland. They found mean values of lead in the runoff of 38 μg/l with a standard deviation of 110 μg/l. Roofing materials are not specified in the article, nor is the lead concentration of the rainfall itself.

To the knowledge of this P.I., Davis and Burns (1999) also contains the first, and only, peer-reviewed study of lead concentrations in runoff from exterior, painted walls. Their work was motivated by a monitoring study of heavy metals in urban runoff performed by Ni et al.(1995) in Prince Georges County, Maryland. In the 1999 paper, Davis and Burns conducted in-depth laboratory studies in which they sprayed synthetic rainwater on exterior, painted building walls. They found concentrations of total lead as high as 28,000 μg/l for surfaces with paint older than 10 years. For this type of paint, the mean value of the concentration was 810 μg/l. Newer paint, 0-5 years old, produced maximum lead concentrations of 370 μg/l and a mean concentrations of 27 μg/l. The authors attribute the difference between these two sets of observations to the fact that the older paint is more likely to contain high amounts of lead, and also that older paint is likely to peel more easily and hence yield more particulate lead. They also found that the more intense the spray, the higher concentrations the concentrations of lead measured.

The potential applicability of the Davis and Burns wall runoff study to the New Orleans area, or any other area with high numbers of homes with leaded painted exteriors, is obvious. Mielke et al.(1999) has demonstrated that much of the New Orleans top-soil has lead concentrations of more than 300 μg/g and that there are sections of the city with soil concentrations as high as 1100 μg/g. Mielke also noted that the largest concentrations of lead were found at the base of the exterior walls. Although not discussed by Mielke et al, this finding can logically be taken to indicate *that wall runoff is responsible for much of the lead found in the soil, even though soil lead is typically attributed to the lingering effects of leaded gasoline auto exhaust.*

This review of the literature indicates that there is emerging evidence that wall runoff from surfaces with leaded paint is an important source of lead in urban runoff. In addition, some evidence exists that rooftops may store lead particles, and release them in particulate or dissolved form during rainfall events. For TMDL purposes, however, more quantitative information is required. And, for the development of watershed and sub-watershed-based TMDLs in the State of Louisaina, characterization of this pollution and its relationship to rainfall *on a site-specific basis* must be undertaken. Development of these relationships should be of particular concern in New Orleans and other areas of the state where leaded-paint surfaces are prevalent.

For these reasons, this project aims to provide models, applicable to New Orleans, which predict lead concentrations from rooftops and exterior leaded-paint walls as functions of building characteristics and rainfall characteristics. These models may also be applicable to other parts of the state and to the country – it is hoped that model validation for these areas will be undertaken as part of a broader study, perhaps sponsored by the LA Department of Environmental Quality, once this proposed study is completed.

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